

### Microbiological, Geochemical and Hydrologic Processes Controlling Uranium Mobility: An Integrated Field-Scale Subsurface Research Challenge Site at Rifle, Colorado

PI: P. E. Long

Co-Pl's: J. Banfield, D.P. Chandler, J.A. Davis, B. Hettich, P.R. Jaffe, L.J. Kerkhof, R.K. Kukkadapu, M. Lipton, A. Peacock, N. VerBerkmoes, K.H. Williams, S.B. Yabusaki

Key collaborators: D. Lovley, D. Holmes, L. N'Guessan, P. Mouser, T. Scheibe, F. Spane, S. Hubbard, C. Steefel, A. Englert, B. Baldwin

Field Site Co-Managers: R. Dayvault and S. Morrison

DOE-LM Site Manager: R. Bush

DOE-SC Program Manager: Todd Anderson

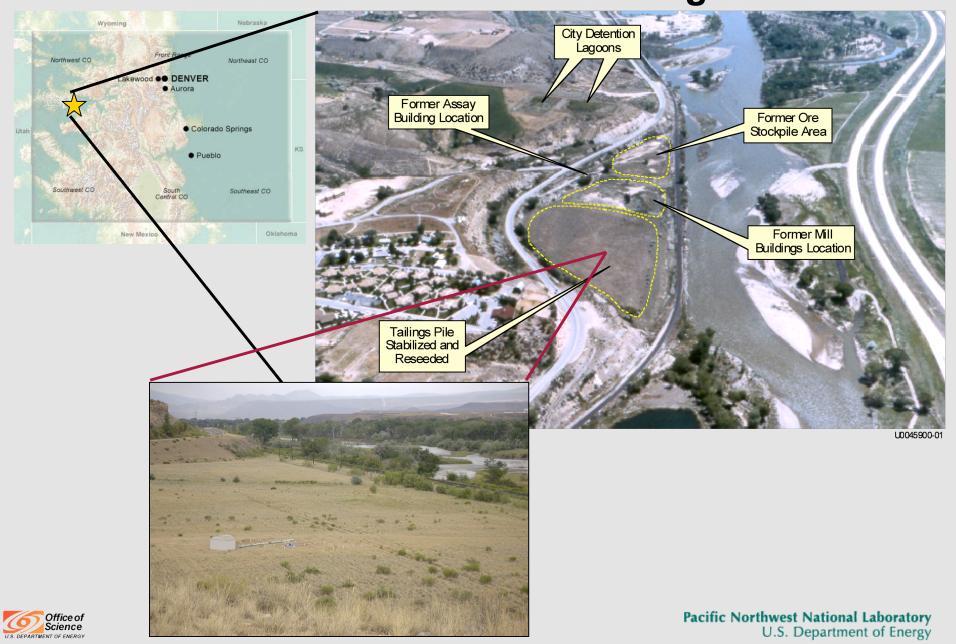
National Laboratory
Operated by Battelle for the
U.S. Department of Energy



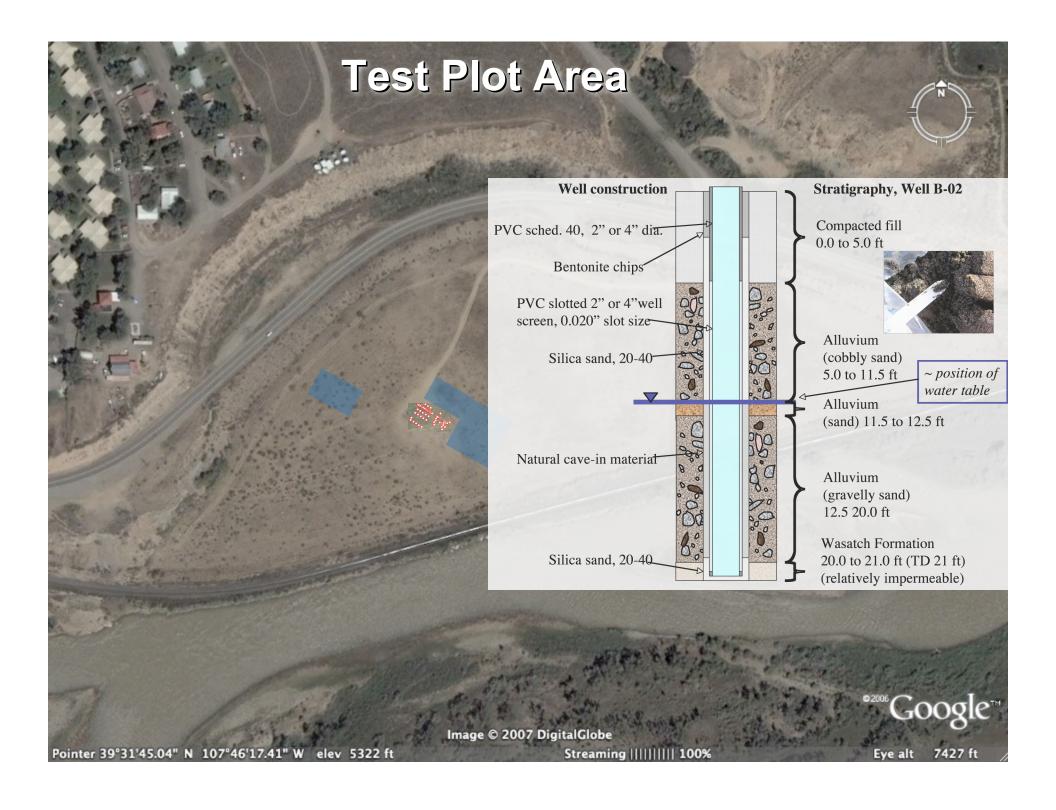
# U.S. Department of Energy: "Problem owner" for a wide range of sites contaminated with metals and radionuclides

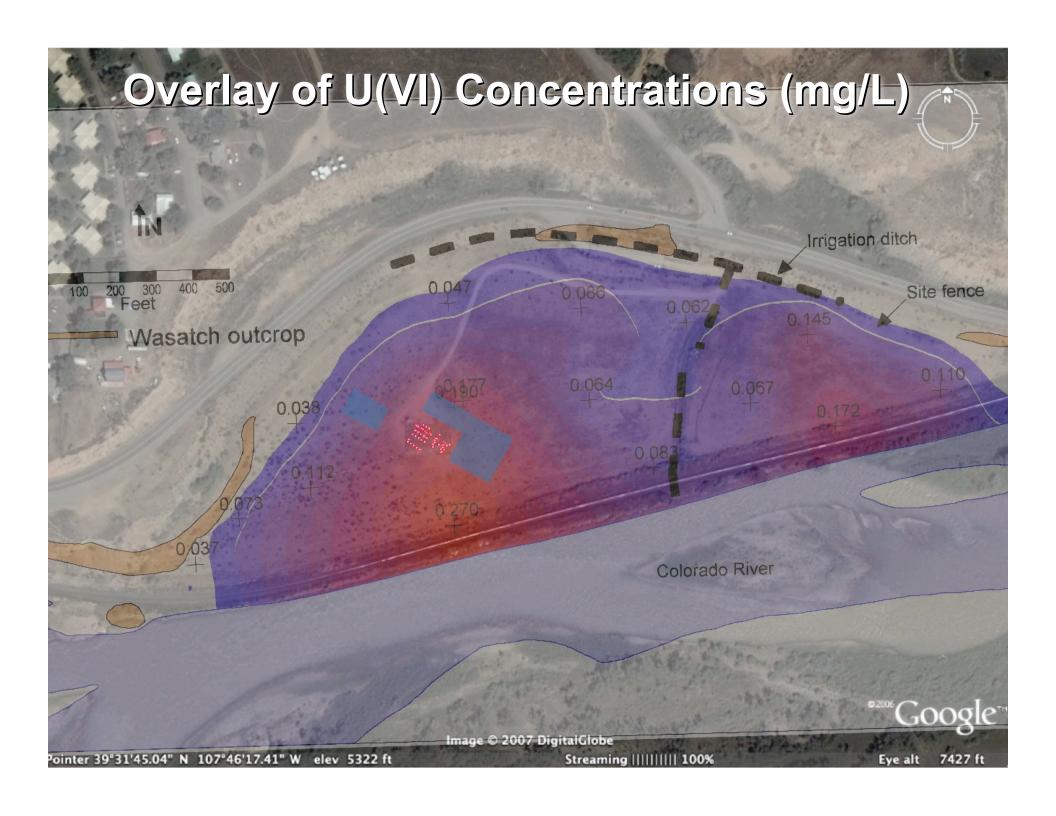


# Context for field bioremediation research at the Old Rifle Uranium Mill Tailings Site





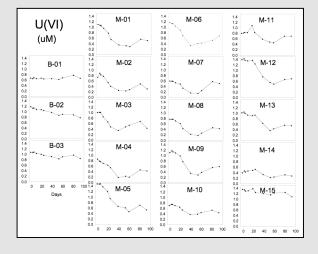


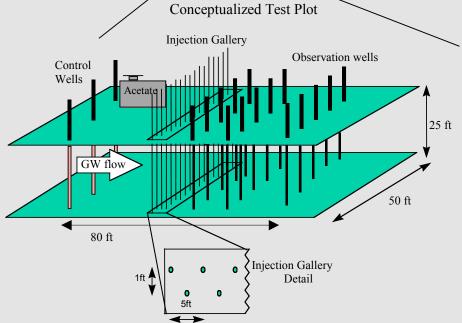


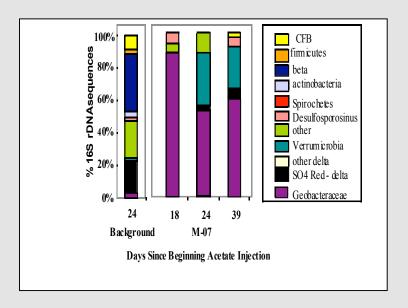
## **Test Plot Design(s)**







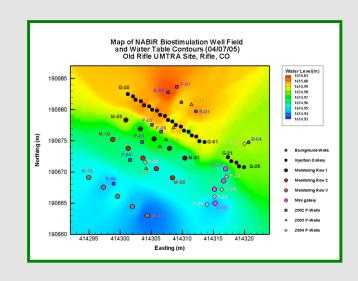


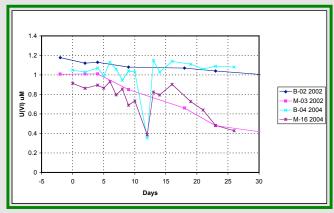




### **Summary of Previous Work**

- Amendment with acetate stimulated metal reduction and loss of soluble uranium from the groundwater
- Loss of soluble uranium correlated with a substantial shift in the subsurface microbial community towards members of the Geobacter family
- Geobacteraceae are known to couple acetate oxidation to the reduction of Fe(III) oxides and U(VI)
- Loss of soluble uranium is attributed to the stimulation of Geoabacteraceae and enzymatic reduction of soluble U(VI) to insoluble U(IV)
- Continued acetate addition caused shift towards sulfate reduction decreased loss of U(VI) from groundwater
- Unexpected removal of U(VI) continued after stopping addition of acetate







#### Underlying constraints on application of in situ bioremediation of uranium

#### Incomplete understanding of:

- Function and growth of subsurface microbial communities
- Microbial community succession in the subsurface
- ▶ Relative importance of enzymatic reduction of U(VI) to other potential immobilization mechanisms:
  - Sorption
  - Mineralization
  - Relative potential for abiotic reduction
- Long-term stability of immobilized uranium
- Methods to monitor stimulated metal reduction and microbial communities in the subsurface



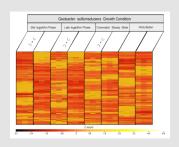
### Objective of the Rifle IFC

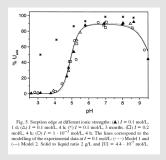
- ▶ Provide a "comprehensive and mechanistic understanding of the microbial factors and associated geochemistry controlling uranium mobility in the subsurface"
  - Such understanding of the coupled biotic and abiotic factors affecting uranium mobility is relevant to DOE's cleanup and long-term stewardship missions

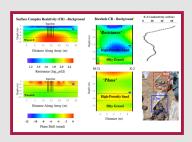


#### **Overall Science Themes**

- Mechanisms of U bioreduction illuminated by protein expression
- ▶ Relative contribution of biotic processes and abiotic uranium immobilization processes evaluated (e.g. U bioreduction and U sorption)
- Correlation of subsurface geochemical processes with geophysical monitoring of subsurface redox status associated with bioreduction
- Comprehensive reactive transport modeling of uranium mobility in the subsurface.









# Hypothesis 1: Extending microbial Fe reduction in the subsurface

- ► HYPOTHESIS 1: In the presence of mM sulfate concentrations in groundwater, the transition from Fe(III) to sulfate reduction during acetate amendment will occur when the readily bioavailable Fe(III) is depleted. Iron reduction (and concomitant U(VI) reduction) can be extended in time through:
  - 1) the addition of nanoparticulate or soluble Fe(III) to the subsurface, and
  - 2) introduction of acetate at concentrations sufficient to support iron-reduction but not sulfate-reduction.



# Hypothesis 2: Impact of reducing conditions on U(VI) sorption

► HYPOTHESIS 2: The sorption of U(VI) under reduced conditions is decreased overall in comparison to more oxic conditions, but is still large enough to retard U(VI) transport in the Rifle aquifer relative to groundwater flow.

Quantifying the impact of U(VI) sorption on groundwater U(VI) concentrations under Fe-reducing conditions is a crucial part of numerical modeling of aquifer conditions during and after biostimulation experiments.



### Hypothesis 3: Long-term postbiostimulation removal of U(VI)

- ► HYPOTHESIS 3: Long-term post-biostimulation removal of U(VI) is dependent on ferrous sulfide minerals precipitated during sulfate reduction. After cessation of acetate amendment, these minerals become electron donors for a *post-biostimulation microbial community* capable of using low ambient concentrations of oxygen and nitrate as terminal electron acceptors. U(VI) is sorbed on to;
  - 1) biopolymers specific to the post-biostimulation microbial consortia, and/or,
  - 2) freshly oxidized Fe(III) mineral surfaces.



# Hypothesis 4: Naturally occurring rates of U(VI) reduction

► HYPOTHESIS 4: Slow, naturally occurring rates of microbially mediated U(VI) reduction can be estimated (low, medium, high) using molecular biomarkers in Rifle samples by comparing the lowest acetate amendment in Hypothesis 1 with samples from other Rifle locations with no electron donor amendment.



### Innovative Approaches and Techniques

- Proteomics
- 13C labeling
- Detailed analysis mineralogic changes,
- Lab and field sorption experiments under reducing conditions
- Geophysical monitoring (especially ERT, complex resisitivity)
- Detailed monitoring of hydraulic conductivity before, during, and after biostimulation
- CATs (cellularly adsorptive tracers)
- Instrumented in situ incubators
- Integration of diverse data sets via joint inversion
- Comprehensive reactive transport modeling

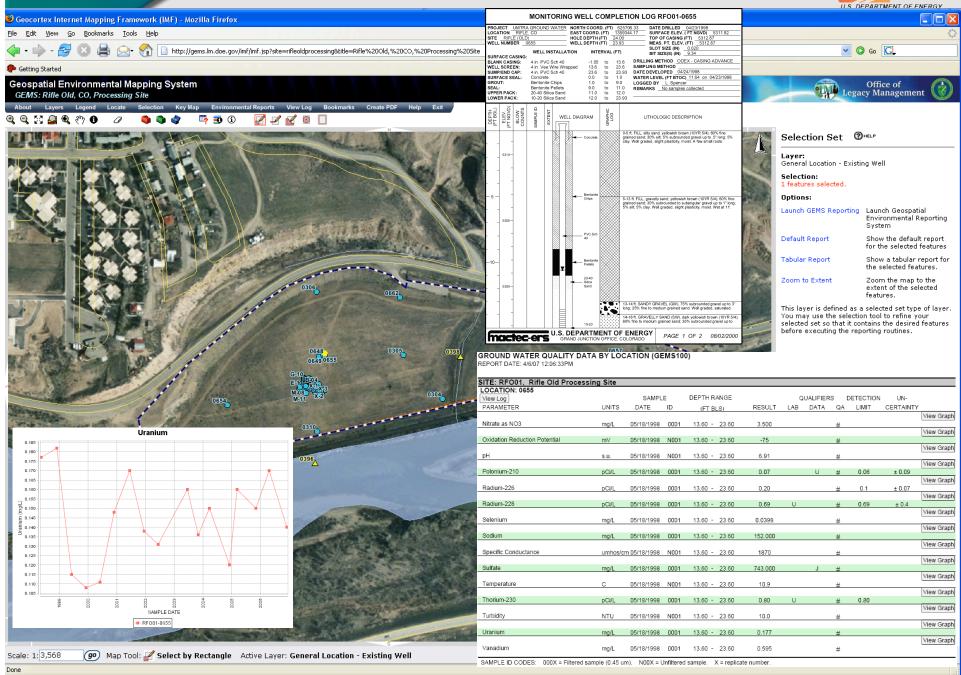


#### FY-07 Schedule

- Kick-off Meeting in Grand Junction, CO Feb 27-Mar 1, 2007
- Mid- to late-May 2007: Electromagnetic survey of entire flood plain
- May 15, 2007: Geophysics and backhoe sampling for assessing experimental site locations
- June 15, 2007: Well installation for 2007 experimental site
- ▶ July 30 to August 15, 2007: Initial field experiment starts
- October 1, 2007: Initial field experiment ends

#### Old Rifle Data Availability on DOE-LM Web Site







#### **Available Site materials**

- Groundwater samples under background and biostimulated conditions
- Filtrates of groundwater samples
- Sediment samples under background and biostimulated conditions
- ► Large volumes of Ucontaminated sieved sediment <2mm (RABS)
- Mineral separates on an asrequested basis (small volumes)









# Opportunities for collaboration or augmentation of existing science team

- Groundwater organic carbon fractions
- Sedimentary structure and architecture
- Sediment geochemical properties
- ► Alternate electron donors
- Sampling techniques that preserve sedimentary structures
- Others...



### Summary

- The four hypotheses are inter-related and elements of all four will be examined in each of the laboratory and field experiments planned for the site
- Please visit the Rifle IFC poster this evening
- Selected Co-PIs will now discuss the key approaches to investigating these hypotheses



#### Rifle IFC Presentations:

- DOE-LM Perspective on science needs and the Rifle IFC. Rich Bush (5 minutes)
- Biogeochemistry and reactive transport modeling of in situ biostimulation experiments at the Rifle site. Steve Yabusaki (20 minutes)
- Rifle IFC geochemistry and abiotic U(VI) reactions under ironreducing conditions. *Jim Davis* (25 minutes)
- Protein measurement for assessment of subsurface microbial activity under biostimulated conditions. *Jill Banfield* (25 minutes)
- Hydrogeophysics and electrical methods for monitoring TEAPs at the Rifle IFC. Ken Williams (25 minutes)



### **Acknowledgements**



- ► Funding: Environmental and Remediation Sciences Program (ERSP), Biological and Environmental Research (BER), Office of Science, U.S. Department of Energy
- ► Thanks for Cooperation:
  - City of Rifle, Colorado
  - Colorado Department of Public Health and Environment
  - U.S. Environmental Protection Agency, Region 8
- Pacific Northwest National Laboratory is operated by Battelle for the United States Department Of Energy under Contract DE-AC06-76RL01830